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Nonequilibrium Superconducting States  
with Two Coexisting Energy Gaps

Gerd Schön (a)

Institut für Theorie der Kondensierten  
Materie, Universität Karlsruhe, BRD

and

André-M. Tremblay

Laboratory of Atomic and Solid State  
Physics, Cornell University, Ithaca  
New York, 14853

The effect of tunnel currents in superconducting junctions on the energy gap is calculated. For certain parameters two different gaps can exist. The stability of these solutions is investigated and at a certain voltage a "first order transition" is found. This result explains the experimentally observed inhomogeneous states in superconducting tunnel junctions.

Recent experiments on superconducting tunnel junctions<sup>(1)(2)</sup> suggest that for certain injection currents and voltages a superconductor sustains simultaneously two different values of the energy gap. Existing phenomenological models<sup>(3)</sup> predict that above a critical density of the excitations  $n_c$ , the superconductor has an intrinsic instability with respect to the formation of a spatially inhomogeneous state. This is not in agreement with the experimental results of Gray and Willemsen<sup>(2)</sup>. These authors interpreted the effect by inhomogeneities in the probes, with a lower gap value, which grow with increasing total current.

In this letter we describe microscopically a superconducting tunnel junction consisting of an injector and a probe and find that two stable values of the energy gap can exist in the probe. In accord with the experiments, at a certain voltage a first order phase transition takes place where the part of the probe with the lower gap and larger injection current density grows while the part with the larger gap and lower injection current density decreases. The relative size of the two regions is controlled by the total injection current. This is analogous to a liquid-gas transition at a certain pressure where the relative volumes are controlled by the total volume. Our result differs from the model of Ref. 2 insofar as we find the existence of two gap values to be an intrinsic property of the system. Furthermore, our approach describes the gap enhancement by quasiparticle extraction, investigated in the experiments of Chi and Clarke<sup>(4)</sup>. We will present a detailed analysis in two limits, near  $T_c$  and near  $T = 0$ . Qualitatively, the same results can be obtained for any intermediate temperature.

We assume that the injector is thick and not appreciably perturbed by the current and that the phonons remain in equilibrium. Thus, the probe can be studied using the kinetic equations for the quasiparticle distribution function and the order parameter derived in Ref. 5. In general

there is no simple relation between the injected current (branch imbalance<sup>(6)</sup>), and the total number of excitations (electron-plus hole-like) created in the probe. Only the latter quantity modifies the magnitude of the order parameter. The corresponding deviation of the distribution function  $\delta f_E$ , which is odd in energy, is obtained from the Boltzmann equation:

$$\mathcal{N}_1(E)\delta f_E - K(\delta f) = P_E - \mathcal{N}_1(E) \cdot \partial f^0(E)/\partial E \cdot \Delta\Delta/E . \quad (1)$$

$\mathcal{N}_1(E)$  is the normalized BCS density of states and  $K(\delta f)$  describes inelastic electron-phonon scattering. It suffices to know that  $K(\delta f)$  can be split into a "scattering out" term  $-\tau_E^{-1} \mathcal{N}_1(E)\delta f_E$ , and a "scattering in" term, an integral operator. The perturbation is  $P_E = B \mathcal{N}_1(E) [\mathcal{N}_1^{-1}(E - eV)(\text{th } E/2T - \text{th } (E - eV)/2T) + (eV \leftrightarrow -eV)]$  where  $B^{-1} = R\Omega e^2 N_0$ ,  $R$  is the resistance of the junction,  $\Omega$  the volume of the probe,  $N_0$  the normal density of states, and  $\mathcal{N}_1^{-1}$  refers to the injector. The thickness of the probe is small, and no spatial variations in this direction occur. For the moment we also neglect spatial variations in the junction plane. The effect of  $\delta f_E$  on  $\Delta$ , the magnitude of the order-parameter, follows for temperatures near the critical temperature of the probe from the Ginzburg-Landau (G-L) equation<sup>(5)</sup>

$$\pi\dot{\Delta}/8T - \chi\Delta = (\alpha - \beta\Delta^2)\Delta . \quad (2)$$

where  $\chi = - \int_{-\infty}^{\infty} dE 1/E \mathcal{N}_1(E) \delta f_E$  and  $\alpha = (T_c - T)/T_c$ ,  $\beta = 7\zeta(3)/8\pi^2 T^2$ .

We obtain analytic results if we assume that the energy gap of the injector and the voltage are small  $\Delta_1/T, |eV|/T \ll 1$ . The linear term of an expansion of  $P_E$  in powers of  $eV/T$  is localized in a narrow energy region  $|E| \leq \Delta_1 + |eV| \ll T$ .

Consequently, the corresponding contribution  $\delta f_E^{(1)} = \tau_0 P_E / V_1(E)$  to the stationary nonequilibrium distribution, is obtained neglecting the "scattering in" term and taking the scattering time  $\tau_E$  at  $E = 0$ . Depending on the relative values of  $\Delta, \Delta_i$  and  $V$ ,  $\delta f_E^{(1)}$  describes an increase or decrease of the density of excitations near the gap edge. The resulting  $\chi^{(1)}(\Delta, V)$  also changes sign, and both gap reduction or gap enhancement are possible. By contrast, the quadratic term of the expansion of  $P_E$  always results in a gap reduction. Since  $\delta f_E^{(2)}$  is not localized near the gap edge, the "scattering in" term cannot be neglected. A variational calculation<sup>(7)</sup> yields  $\chi^{(2)} = -1.4 B \tau_0 (eV/2T)^2$ . This contribution can be interpreted as an effective increase in temperature. For the following discussion we shall neglect it, although it may be important for a quantitative analysis.

In the considered limit ( $\Delta, \Delta_i, |eV| \ll T$ ), we find

$$\chi^{(1)}(\Delta, V) = -2\theta(\Delta + \Delta_i - |eV|) B \tau_0 |eV/2T| g((d - |eV|) K(k) + (c - d) \pi(a^2, k)) \quad (3)$$

where  $K$  and  $\pi$  are complete elliptic integrals of the first and third kind,  $g = 2/[(a - c)(b - d)]^{1/2}$ ,  $a^2 = (b - c)/(b - d)$ ,  $k^2 = a^2(a - d)/(a - c)$ , and  $a, b, c, d$  are the parameters  $|eV| \pm \Delta_i$  and  $\pm \Delta$  assigned such that  $a > b > c > d$ . In order to obtain the stationary solution of the G-L equation including the non-equilibrium term, we employ graphical constructions. The intersections of  $-\chi^{(1)}(\Delta, V)$  and the curve  $a - \beta \Delta^2$  yield possible solutions of  $\Delta$ . A large positive value of  $\chi^{(1)}(\Delta, V)$  at  $|eV| = \Delta_i - \Delta$  (for  $\Delta_i > \Delta$ ), corresponding to a net extraction of excitations, results in a large gap enhancement at this voltage. This is in qualitative agreement with the experimental results of Chi and Clarke<sup>(4)</sup>.

For our present problem, larger voltages are of interest. As shown in Fig. 1,  $\chi^{(1)}(\Delta, V)$  has a step structure at  $|eV| = \Delta_i + \Delta$ . This step corresponds to the step in the

$I(V)$  characteristic of an ideal SIS tunnel junction at the same voltage, however,  $\chi^{(1)}$  is finite at low voltages  $|eV| \leq \Delta + \Delta_i$  and zero above. In the vicinity of the step, we can approximate Eq.(3) by

$$\chi^{(1)}(\Delta, V) = B \tau_0 |eV/2T| \pi \sqrt{\Delta_i/\Delta} \Theta(\Delta + \Delta_i - |eV|). \quad (4)$$

For suitably chosen parameters (e.g.,  $eV = 2.2\Delta_i$  in Fig.1) we find three solutions of the G-L equation denoted by  $\Delta_1$ ,  $\Delta_2$ ,  $\Delta_3$ . (In the presence of any level broadening, the step actually has to be replaced by a finite slope, therefore,  $\Delta_2$  is also a solution.) In addition  $\Delta = 0$  is a solution. From an analysis of the time dependent equations, we find that  $\Delta = 0$  and  $\Delta_2$  are unstable, whereas  $\Delta_1$  and  $\Delta_3$  are locally stable. At low voltages, we find only one enhanced superconducting solution, whereas at high voltages we find only the unperturbed solution. In the inset of Fig.1, the solutions  $\Delta$  as a function of the voltage are shown. Obviously, in the range where  $\Delta(V)$  has two values with  $\Delta_i + \Delta_1 > |eV| > \Delta_i + \Delta_3$ , two significantly differing values of the current density are obtained.

In order to find which of the two locally stable solutions is globally stable, we follow the analysis performed by Schmid<sup>(8)</sup>. The probability of a solution  $\Delta$  is given by  $W(\Delta) \propto \exp(-\tilde{F}(\Delta)/T)$ , where the generalized free energy is  $\tilde{F}(\Delta) = -2N_0 \Omega \int_0^\Delta d\Delta' [\alpha - \beta \Delta'^2 + \chi(\Delta')] \Delta'$ .  $\tilde{F}$  is

plotted in the inset of Fig. 1. The situation is clearly analogous to a first order phase transition. At low voltages,  $\Delta_1$  is the globally stable "phase". At a certain voltage  $V_o$ , where  $\Delta_i + \Delta_1 > |eV_o| > \Delta_i + \Delta_3$ , we have  $\tilde{F}(\Delta_1) = \tilde{F}(\Delta_3)$ , and a transition between  $\Delta_1$  and  $\Delta_3$  can take place. With increasing total current, the size of the region in the  $\Delta_1$  "phase" with small injection current density decreases in favour of the region in the  $\Delta_3$  "phase" with large injection current density. At high

enough voltages, the  $\Delta_3$  "phase" is globally stable. In addition, the metastable states will result in a hysteresis under suitable conditions. The effect of  $\chi^{(2)}$  or generally of heating will be to reduce  $\Delta_1$  and  $\Delta_3$  below the values found here, but these processes are "smooth" enough that the difference  $\Delta_1 - \Delta_3$  will be preserved.

All of these results, including the dependence of  $\Delta_1 - \Delta_3$  on the junction resistance, are in good agreement with the experiments. Even the hysteresis along the lower branch of  $I(V)$  has been detected. In the case of stimulation of superconductivity by microwaves or by phonons at a certain temperature two stable solutions are found too<sup>(8)</sup> (normal and superconducting or both superconducting with different gaps, respectively). However, the transition between these solutions occurs abruptly, since there is no external variable, such as the total injection current in the present problem to control the transition.

Even in the state where the two gaps coexist, the distribution function  $\delta f_E$  (in the limit considered) is single valued. Therefore, spatially inhomogeneous problems are described by adding the space derivatives to the G-L equation. We find<sup>(9)(10)</sup> that at  $V = V_o$  a stationary wall separating the two phases can exist. Apart from the fact that the location of the wall can be shifted arbitrarily, this solution is locally stable. This confirms our result of the stable coexistence of the two phases at the voltage  $V = V_o$ . On the other hand, droplets or periodic structures correspond to saddlepoint solutions and are therefore unstable.

In the low temperature limit ( $\Delta, \Delta_1 \gg T$ ) qualitatively similar results can be found. Particularly interesting are injection voltages close to the sum of the two gaps:  $|(\Delta_1 + \Delta) - |eV|| / \Delta \ll 1$ . In this case  $P_E$  and consequently  $\delta f_E$  are localized near the gap edge. Integrating the Boltzmann equation with respect to the energy, we find an equation for the total number of excitations per unit

volume  $n = 4N_0 \int_0^\infty dE N_1(E) \delta f_E$  in the form of a linear Rothwarf-Taylor equation<sup>(11)</sup>. In the stationary case,

$$2n/\tau_R(E = \Delta) = 4N_0 \int_0^\infty dE P_E = |I(V)/e\Omega|. \quad (5)$$

In deriving Eq. 5, the localization of  $\delta f_E$  allowed us to neglect the energy dependence of the recombination rate and take its value at the gap edge<sup>(12)</sup>  $\tau_R^{-1}(E = \Delta) = 6.5\tau_0^{-1}(T/\Delta)^{1/2} \exp(-\Delta/T)$ . Furthermore, within terms of order  $\exp(-\Delta/T)$ ,  $\int_0^\infty dE P_E$  is proportional to the injection current. The effect of nonequilibrium excitations on the order parameter follows from the self-consistency relation  $\Delta = \lambda \Delta \int_0^{\Delta_0} dE 1/E N_1(E) (\ln E/2T - 2\delta f_E)$ . Again the localization of  $\delta f_E$  allows us to simplify this equation by setting  $\Delta/E \delta f_E \approx \delta f_E$ . Thus, to lowest order in  $\epsilon = (\Delta - \Delta_0)/\Delta_0$ , where  $\Delta_0$  is the unperturbed  $T = 0$  gap, we obtain

$$\epsilon = -n/2N_0 \Delta_0 \theta((|eV| - \Delta_0 - \Delta_i)/\Delta_0 - \epsilon). \quad (6)$$

The step in the injection current at the voltage  $|eV| = \Delta + \Delta_i$  leads to a corresponding step in  $n$ , which is made explicit in Eq. (6). For suitably chosen parameters, we find again two locally stable solutions  $\epsilon = 0$  and  $\epsilon = -n/2N_0 \Delta_0$ . The physical interpretation of the two solutions is straightforward. First we notice that near  $T = 0$  there are no excitations which could be extracted from the system, and an injected current increases their number. At a voltage  $|eV|$  slightly below  $\Delta_i + \Delta_0$ , the probe can be either in the unperturbed gap state  $\Delta_0$  with no current or in a low gap state with finite injection current and consequently increased excitation number stabilizing the low gap value.

At any intermediate temperature  $0 \leq T \leq T_c$ , the strong voltage dependence of the tunneling current at  $|eV| \approx \Delta_1 + \Delta$  results in a steplike modification of the quasiparticle distribution. For this reason we expect the qualitative result of this paper, the coexistence of two different gap values, to be true at all temperatures below  $T_c$ .

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Figure Caption

Fig. 1: Graphical solution of the G-L equation for voltages  $|eV| > \Delta_i$ . The constants  $\alpha, \beta$  and  $B$  in  $\chi^{(1)}$  are chosen arbitrarily, the higher order term  $\chi^{(2)}$  is neglected. The unperturbed solution is chosen to be  $\Delta_0 = \Delta_i$ .  $\chi^{(1)}(\Delta, V)$  has a step at  $\Delta = |eV| - \Delta_i$ . For the same parameters, the generalized free energy  $\mathcal{F}(\Delta, V)$  is shown in the upper inset, while the solutions  $\Delta(V)$  are shown in the lower inset.

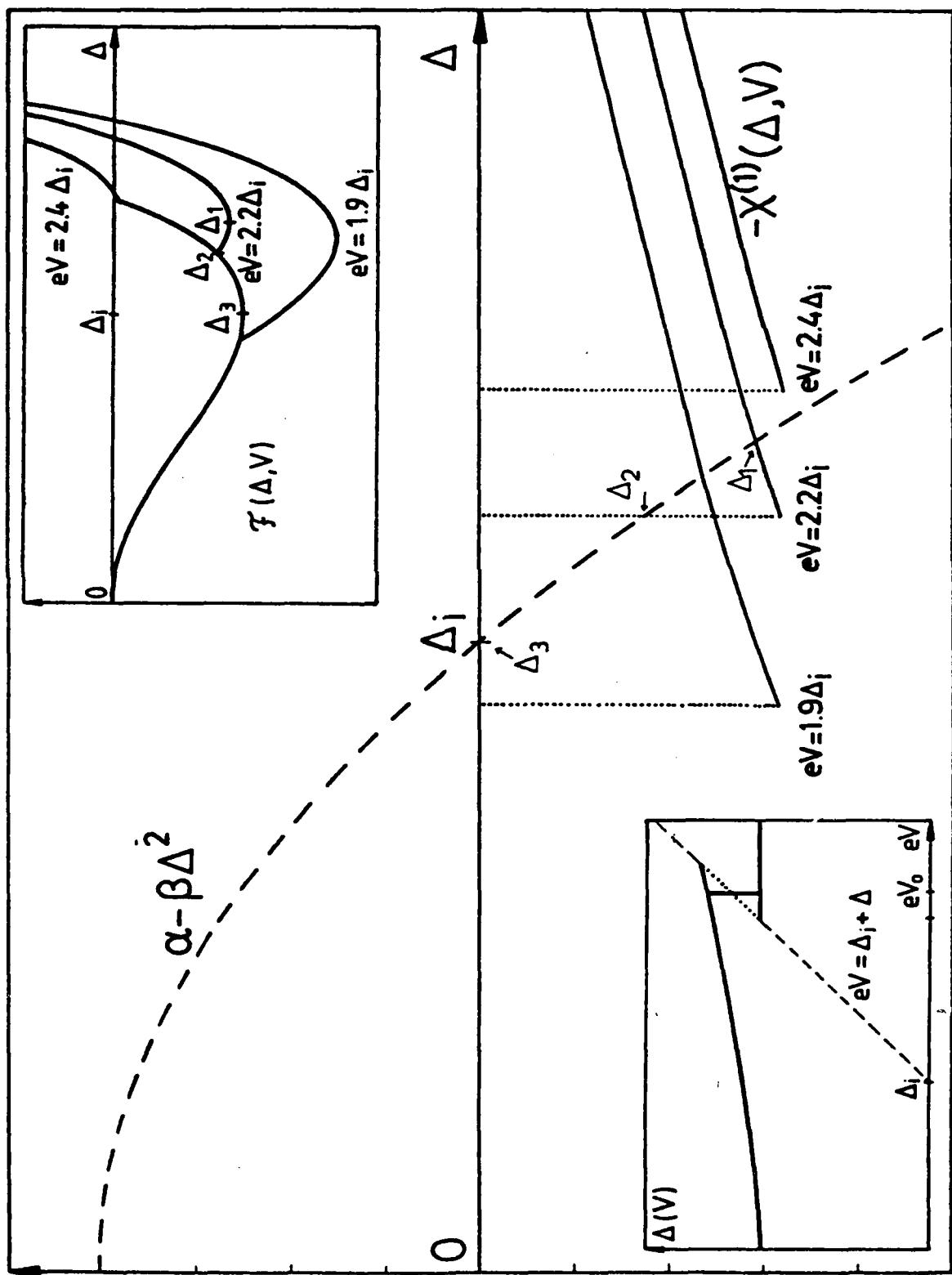


Fig. 1

